Schofield, R. K. and Blair, G. W. S. 1932. Plastic strength of soft materials, the relationship between viscosity, elasticity and plastic strength of soft materials as illustrated by some mechanical. *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences.* 138 (836), pp. 707-718.

criticism, and to the Ordnance Committee and to the Commandant of the Military College of Science for permitting publication.

Summary.

A method of analysing pressure-time curves obtained from closed-vessel experiments is developed, whereby two functions of the pressure are plotted against time. One yields a straight line if the rate of burning is proportional to the density, while the graph of the other is straight if the rate is proportional to the pressure. Only one of the four propellants considered by Crow and Grimshaw ('Phil. Trans.,' A, vol. 691 (1932)) gives evidence of a departure from the pressure law.

The Relationship between Viscosity, Elasticity and Plastic Strength of Soft Materials as Illustrated by some Mechanical Properties of Flour Doughs, I.

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[Plate 15.]

1. Flour dough belongs to a group of materials in which a high degree of plasticity is combined with considerable elasticity. Owing to their great industrial importance, a number of technological investigations have been carried out on these materials (of which unvulcanised rubber is another important example); but the problem of bringing the description of their behaviour within the scope of a general theory of viscosity and elasticity has hardly been tackled.

The time during which a stress is applied is as important as the magnitude of the stress in determining how much of the deformation is elastic (recoverable) and how much plastic (non-recoverable). This fact suggests that a formulation based on Maxwell's "time of relaxation" should be of value in this connection. The formulation as Maxwell gave it applies to a true fluid, for which case the relaxation is exponential and for which the rate of dissipation of internal stress is proportional to the stress, the constant of proportionality being the reciprocal of the relaxation time. Thus

$$-\frac{ds}{dt} = \frac{1}{t_r}\,S,$$

or

$$\frac{1}{t_r} = -\frac{1}{S}\frac{dS}{dt} = -\frac{d\,(\log_e S)}{dt},$$

where S is the shearing stress, and t_r the relaxation time.

An obvious method of extending this conception to plastic materials is to recognise the relaxation time as a quantity which is not a constant but which varies according to the stress, and to consider its reciprocal as determined by $-d(\log_e S)/dt$. Thus the reaction of the material to an external stress will be more purely elastic in proportion as the time of relaxation exceeds the time of application.

In handling a flour dough, strains up to 30 per cent. given momentarily recover almost completely, so that, for the corresponding stresses, the time of relaxation must be large enough to be measured without the aid of elaborate apparatus. The reason for this is brought out by a further consideration of Maxwell's theory. In steady flow the velocity gradient (or rate of change of shearing strain), G, is related to the rate of dissipation of internal stress by the rigidity modulus, n, of the material. Thus

$$-\frac{dS}{dt} = nG.$$

So that the viscosity, η, which is defined as the ratio of S to G is given by

$$\eta = nt_s. \qquad (1)$$

Hence

$$t_r = \eta/n.$$

In the case of plastic materials, η, if defined as the ratio of S to G, is not a constant,* but varies with S ; n, on the other hand, appears to be a constant of

* There is no universally recognised definition of viscosity in systems other than true fluids. In certain cases, although S/G varies, dS/dG is constant over a considerable range of stress. In such cases the behaviour of the system is conveniently described in terms of dS/dG which has been termed the "pseudo-viscosity"† or "stiffness," its reciprocal being Bingham's "mobility."‡ It seems best to reserve the term viscosity for the simple ratio S/G, and recognise that, in general, this varies with S.

† Scott Blair and Crowther, 'J. Phys. Chem.,' vol. 33, p. 321 (1929).

‡ Bingham, "Fluidity and Plasticity" (1922).

the material. Under low stresses the value of η for flour dough is considerable, while the rigidity modulus is extremely small by comparison with that of ordinary solids. It is the combination of these extreme properties which gives this material a special interest, and makes its study valuable for the purpose of advancing our general understanding of these systems.

2. Experiments which have enabled a direct determination to be made of t_r are dealt with in Section 3. It is convenient, however, before describing these to outline the results of some preliminary experiments which bring out the importance of the time of application in determining the type of reaction to an external stress. The first experiments were carried out with the pachimeter, an instrument for measuring plastic strength under rolling which has already been described in detail in its application to the study of soils.* A test piece in the form of a small cylinder of known radius and length is made to roll between two horizontal plates by reciprocating the lower one, and the instrument is designed to measure the stress which must be exerted by the upper plate in order to produce in the rolling cylinder a permanent elongation.

It was thought that the plastic strength of a dough thus determined might have an important bearing on the baking quality of the flour. It was found that the instrument is capable of distinguishing between flours, but the results did not run entirely parallel with the bakers' opinion. It is now clear that at least one reason for this is that, while the stresses set up by the gas generated in a yeasted dough operate over a time to be measured in hours, the reciprocating action in the pachimeter impresses a stress on any given diameter of the rolling dough cylinder for only a fraction of a second. The fact that a dough cylinder does not show a measurable permanent elongation until a stress greater than a certain critical stress has been applied between the plates must be interpreted as showing that below this critical stress the time of relaxation is too long for any measurable plastic deformation to occur in the time allowed by the instrument. The remarkable reproducibility of the figures given by this instrument is evidently due, in part, to the fixing of the time of application of the stress by the speed of reciprocation ; but it must also, in part, be attributed to the rolling action, through which every diameter comes under stress in turn thereby averaging out the effects of irregularities in the specimen. Iti s evident, however, that for such a material the terms *plastic strength*, and *yield value*, if used to describe the figures obtained, must be construed as

* Schofield and Scott Blair, 'J. Agric. Sci.,' vol. 132, p. 135 (1932) ; 'J. Soc. Chem. Ind.,' vol. 51, p. 205 (1932) ; 'Trans. Ceram. Soc.,' vol. 31, p. 79 (1932).

relative rather than absolute terms. A second series of measurements was made on a newly designed instrument which we have called a rack owing to its superficial resemblance to an ancient instrument of torture, fig. 8, Plate 15. Long cylindrical pieces of dough (made by forcing the material through a short piece of metal tubing attached to the body of a grease gun) were stretched out and held stretched for a measured time, at the end of which they were cut loose and their elastic contraction measured.*

Fig. 1 shows the result of a series of experiments in which cylinders of a dough† of a good bakers' mixture were stretched to different extents and cut loose after 1 minute. It will be seen that the elastic recovery, expressed as

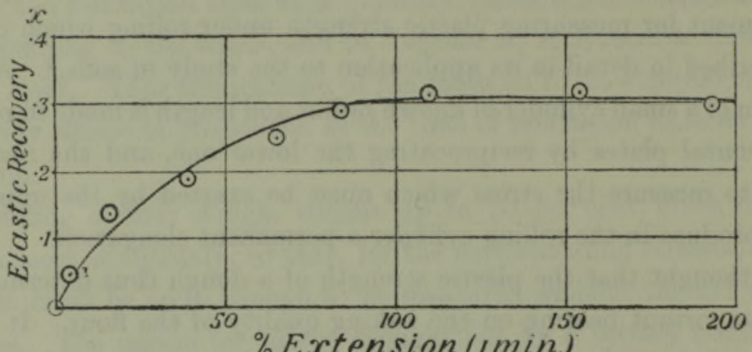

FIG. 1.

the ratio of the contraction to the final length, increased as the initial extension was increased to 100 per cent., but that no further increase was obtained up to 200 per cent. A curve resembling the earlier portion would be obtained on plotting the same quantities for a copper wire as may be seen from the data recently published by Taylor and Quinney.‡ It is not suggested that the correspondence is complete, but a due recognition of the widely different relaxation times involved in the two cases does much to bridge the gap. In a further series of measurements cylinders made from portions broken from a large dough at intervals during a period of several hours were stretched to

* The satisfactory agreement obtained between duplicate dough cylinders made up by this method demonstrates that any irregular strains set up in the dough in the grease gun are not great enough to upset the behaviour of the cylinder when under stress.

† In comparing doughs made from different flours it was not convenient to use in every case the same proportion of flour to water. It was found possible to reproduce dough samples most satisfactorily by making them up to that moisture content at which they will just not stick to a glass plate when pressed firmly on to its surface. All doughs were made up with 23 per cent. salt solution.

‡ 'Phil. Trans.,' A, vol. 230, p. 323 (1931).

just over 100 per cent. and their elastic recovery after 1 minute was measured. No appreciable differences were found. These results are in striking contrast to those obtained from portions of the same dough and investigated in the pachimeter. As will be seen from fig. 2 the pachimeter-values (W) show

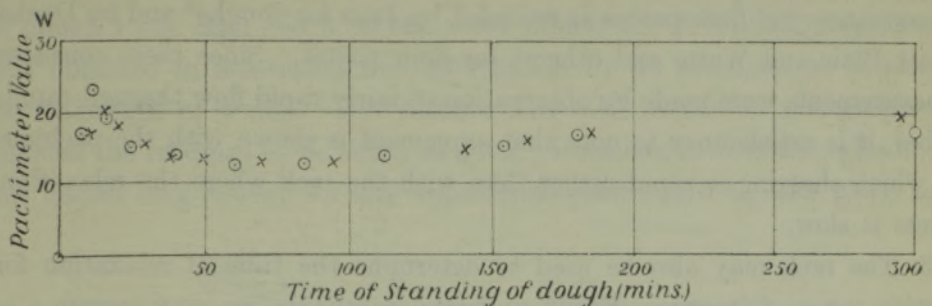

FIG. 2.—⊙ × duplicate doughs.

marked fluctuations with time. In order to ascertain whether the results from the two instruments differ because of a relative shortness of time under stress in the pachimeter, a simple experiment was made. The apparatus constructed consisted essentially of a ballistic pendulum which, being released from an angle of 25°, was allowed, when at the lowest point of its swing, to strike and rebound from the circular face of a cylinder of dough. The cylinder had a radius of 3 mm. and length 15 mm. and was held in an L-shaped support so that the end away from the pendulum remained rigid and the cylinder was given support from below for about three-quarters of its length. The pendulum bob consisted of a clay marble weighing 2·3 gm. supported by two threads

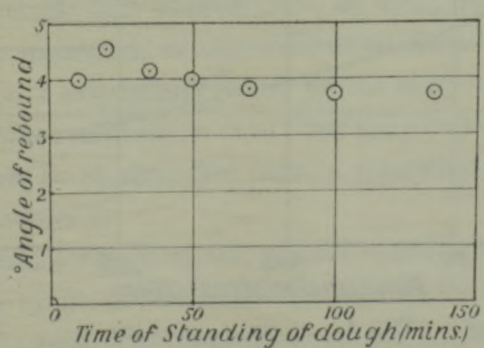

FIG. 3.

45 cm. long separated by 15 cm. at their upper end. The results, shown in fig. 3 (each point represents a mean of three readings) confirm the expectation that the larger values of W, since they indicate a smaller dissipation of rapidly applied stresses, correspond to bigger angles of rebound.

Although this is not the place to discuss how far the variations in W reflect the colloidal and other changes which take place between the time when a baker first mixes his dough and that when it is " ripe " for baking, it is interesting to note that there is a correspondence between them and the variation of consistency of flour pastes as recorded by Jago for doughs* and by Denham, Scott Blair and Watts and others† for flour pastes. Since these consistency measurements were made by observation of fairly rapid flow through capillary tubes, it is satisfactory to note that agreement is shown with the pachimeter in which shearing is rapid rather than with the rack where the relaxation of stress is slow.

3. The rack may also be used to determine the time of relaxation for a certain range of stresses. For if, as seems reasonable, we may assume a constant proportionality to exist between the elastic recovery, x, and the stress still undissipated at the time of cutting loose, the reciprocal of the time of relaxation is as well given by $-d(\log_e x)/dt$ as by $-d(\log_e s)/dt$.‡ The former quantity can be evaluated by obtaining from a series of experiments, in which the initial deformation is the same, the variation of x with the time during which the cylinder is held deformed. The result of such a series is shown in fig. 4, the circles giving the values of x and the crosses of $\log_e x$. Fig. 5 gives

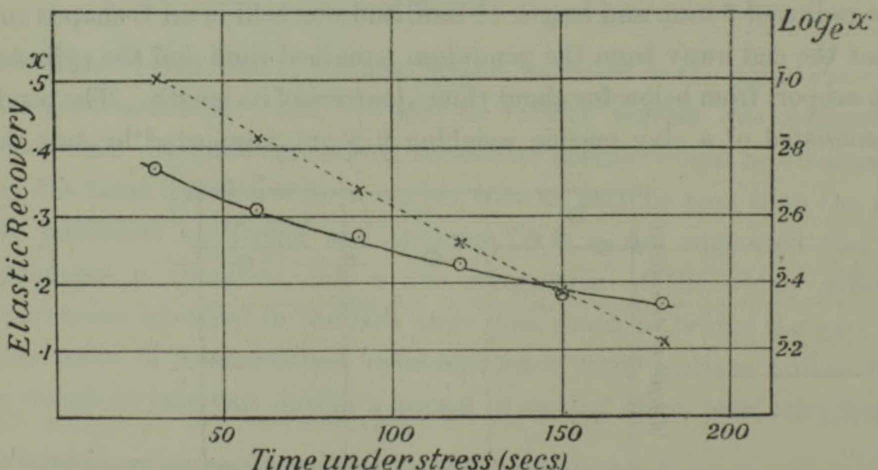

FIG. 4. $\odot = x$, $\times = \log_e x$.

* Jago, "Chemistry of Wheat, Flour, and Bread, and Technology of Bread Making" (1886 and later editions).

† Denham, Scott Blair and Watts, 'Cereal Chem.,' vol. 4, p. 206 (1927); Sharp and Gortner, 'Tech. Bull. Minn. Agr. Exp. Sta., No. 19,' (1923); St. John and Bailey, 'Cereal Chem.,' vol. 4, p. 140 (1929).

‡ Where s is the tensile stress per unit area (equals 3S, see below), the stress plotted in figs. 5, 6 and 7 is the tensile stress, s.

the variation of the time of relaxation with the stress. In order to deduce the undissipated stress plotted as abscissa it was necessary to obtain the appropriate value for Young's modulus. For this purpose observations were made on the compression caused by placing a small weight on top of a squat cylinder of dough 1 cm. high and 0·55 cm. mean diameter. A fourfold magnification was obtained in measuring the deformation by an arrangement resembling the backsight of a rifle. This method was used because it was simple, and enabled the readings to be taken in quick succession, so that the cylinder was not loaded long enough for any appreciable plastic flow to take place, fig. 9,

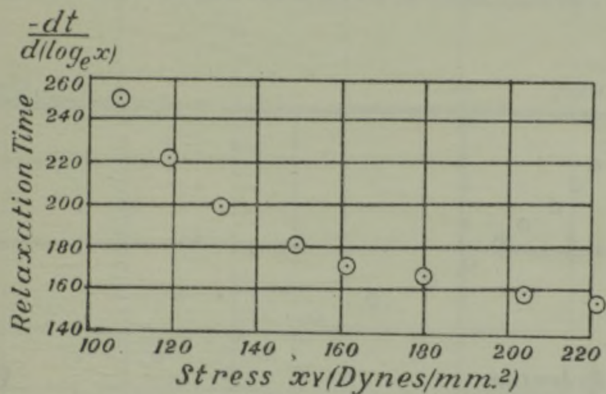

FIG. 5.

Plate 15. A mean value of about 4×10^4 dynes per square centimetre was obtained, and no variations could be detected during the ageing of a dough. This figure is admittedly only approximate, as, apart from the smallness of the deformation to be measured, it was difficult to form the dough into a true cylinder and obtain a satisfactory value for its cross-section. On the other hand, it is certainly not seriously in error; but since higher values were obtained by another method, to be described later, a somewhat higher value, namely, 6×10^4 has been adopted.

In order to obtain a check on the general correctness of fig. 5 and also to extend the stress range of the determination of the time of relaxation, a modification of the rack was constructed in which the weight of the dough cylinder was supported on a pool of mercury contained in a long shallow wooden trough. One end of the dough was fixed by pressing it round a screw let into one end of the trough while the other was attached to a thin strand of rubber by the extension of which the stress on the dough could be observed. The other end of the rubber strand was secured to a thread which could be wound up on a small winch. Direct observations on the decay of the stress were made by first rapidly extending the dough by winding the thread on to the winch until after an extension of about 150 per cent. an ink mark on the dough came into the field of view of a low-power microscope. The thread was then gradually released at rate carefully regulated so as to keep the ink mark on the cross-wire of the microscope. From a separate calibration the stress corresponding to each notch was known, and this, divided by the mean cross-section, gave the ordinate of fig. 6, the abscissa being the recorded times. In fig. 7 is given

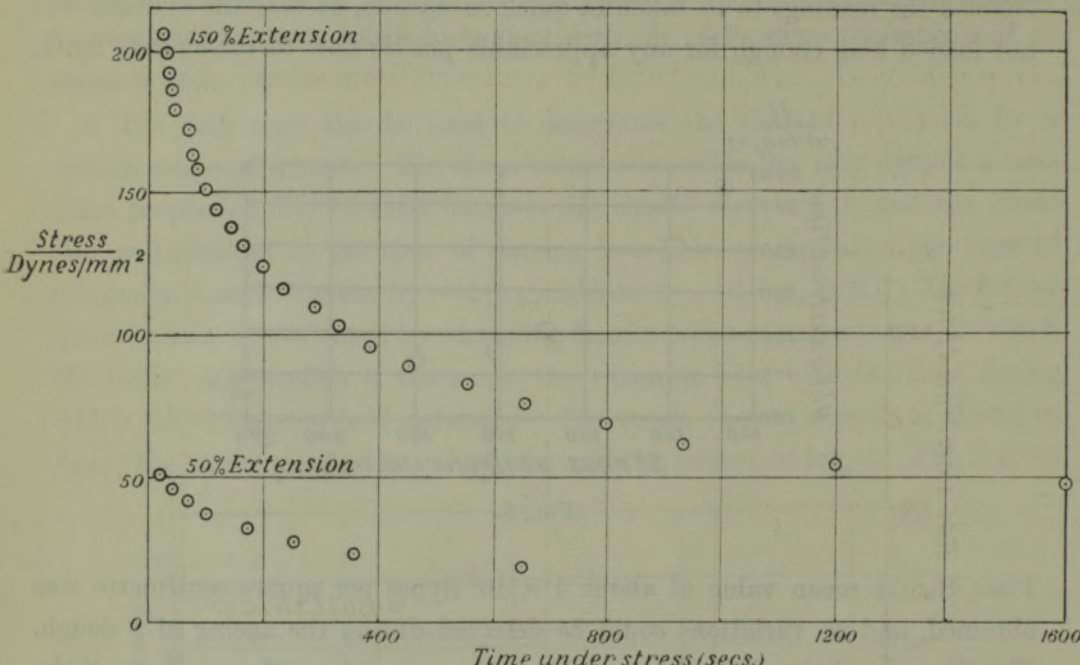

Fig. 6.

the curve for the relaxation time derived from it. It shares with fig. 5 the uncertainty attached to the evaluation of a cross-section, but is in other respects more reliable and extends over a much wider range.

In figs. 6 and 7 are also shown curves relating to the decay of stress for an initial deformation of only 50 per cent. The lack of agreement between the two curves in fig. 7 shows that the time of relaxation depends on the deformation the specimen has suffered as well as on the stress. The dependence of time of relaxation (and hence of viscosity) on the total deformation was suspected from the results of some rougher comparisons of a similar nature made with the original rack. It has been confirmed in a new series of experiments in which observations have been made of the plastic extension of dough cylinders under their own weight, an account of which will be published shortly.

In these experiments it has been found that although the cylinders thin considerably in extending there is no corresponding increase in the rate of extension. This phenomenon may be compared on the one hand with the hardening of metals under working and on the other with an analogous effect observed by Trouton in the case of pitch.* These results point out a weakness† in the usual methods of measuring viscosity by observations on steady flow. Unless special care is taken the only value obtained for the viscosity will be the limiting value for large total deformations.

In a complete study of the plastic behaviour of such a material the viscosity-stress relationship should be found for a series of deformations. The measure-

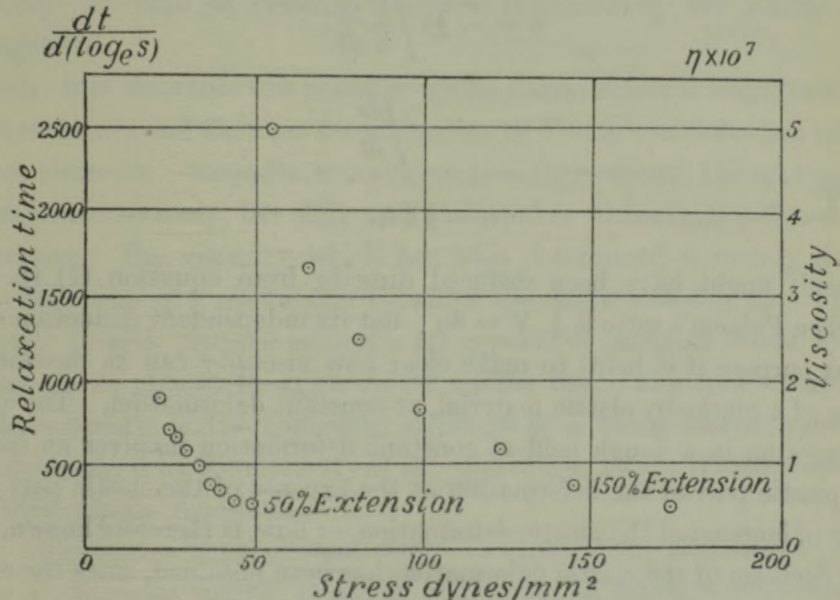

FIG. 7.

ment of flow at constant deformation may seem at first sight to be impossible, but this is actually what was done in the experiments just described. While the winch remained on a given notch the dough was flowing slowly under a substantially constant stress. Before it had gone far, however, the stress was reduced and an elastic recovery occurred equal and opposite to the deformation

* 'Proc. Roy. Soc.,' A, vol. 77, p. 426 (1906).

† The determination of viscosity by the usual methods is also liable to disturbance from slip or anomalous flow at the solid surfaces of the viscometer (Schofield, R. K., and Scott Blair, G. W.,' J. Phys. Chem.,' vol. 34, p. 248, 1505 (1930); 'J. Phys. Chem.,' vol. 35, p. 1212 (1931); 'Phil. Mag.,' vol. 72, p. 890 (1931)). In the method here described no such complication can arise.

caused by the flow. This deformation can therefore be obtained by dividing the stress change by Young's modulus.

According to well-known principles a material such as this for which Poisson's ratio may be taken as $\frac{1}{2}$, when elongated may be considered as subject to two shearing strains each equal in magnitude to the fractional elongation. The corresponding shearing stresses are each one-third of the normal tensile stresses applied in causing the elongation, so that by plotting the rate of shear $-\frac{1}{Y}\frac{ds}{dt}$ against the shearing stress, $\frac{1}{3}s$, a flow-curve is obtained. The viscosity corresponding to any given stress s is therefore given by

$$\eta = -\tfrac{1}{3}s \Big/ \frac{1}{Y}\frac{ds}{dt},$$

but

$$t_r = -s \Big/ \frac{ds}{dt}.$$

Therefore

$$\eta = \tfrac{1}{3}Y\,t_r.$$

This result might have been deduced directly from equation (1) (p. 708), since when Poisson's ratio is $\frac{1}{2}$, $Y = 3n$; but its independent deduction serves a useful purpose if it helps to make clear how viscosity can be measured, in the case of a markedly elastic material, at constant deformation. The change which goes on in a dough held at constant deformation involves an increase of the plastic part of the deformation at the expense of the elastic part; and the rate of increase of the plastic deformation, or flow, is therefore known, if the rate of decrease of the elastic deformation has been obtained, since the sum of the deformations remains constant.

As a check on the value of the Young's modulus, and to test the correctness of using it to relate the undissipated stress to the elastic recovery, an experiment, which was started in the same way as those already described, was carried out on the modified rack. After keeping the ink mark on the cross-wire of the microscope for about a minute, the stress was completely released and the elastic recovery observed. The magnitude of the stress just before it was released having also been observed, it was only necessary to obtain the mean cross-section in order to evaluate Y. The elongation of the cylinders when floated on mercury is not quite so even as when they are supported on rubber bands, and so the cross-section could not be determined very exactly. A value of 6×10^4 was obtained. It has been preferred to the value 4×10^4 obtained with the earlier method as any unevenness in the faces of the loaded cylinder would cause an apparent increase in the deformation thereby giving too low a value for Y. It is to be understood, however, that this value is provisional in view of the difficulties of the experiment. The precise numerical value does not affect the general form of fig. 5.

Discussion.

4. In the foregoing sections it has been shown how the conception of the time of relaxation, which was used by Maxwell as a method of describing the viscous behaviour of a liquid, can be extended to cover the behaviour of a complex system such as a flour dough. An attempt will now be made to specify the type of internal structure indicated by the results of the investigation.

Firstly, it is clear that the dough contains elastic elements which form a connected structure and that the determination of Young's modulus has reference to these elements. Secondly, it is evident that the elements, though connected, are not joined securely, but slide past one another whenever a sufficient stress is operative. The viscosity which has been determined is mainly governed by the behaviour of a plastic film by which the elastic elements are connected. It is quite possible that the elements are capable of complete elastic recovery, but there is at present no criterion for testing this. The time of relaxation is a characteristic of the connected structure as a whole, and its value is as much determined by the elasticity of the elements as by the viscosity associated with their plastic junctions.

The considerable time (several minutes) which often elapses between the release of the stress and the cessation of contraction indicates the existence of another viscosity* which must be distinguished from that already considered. The fact that the dough exhibits elastic recovery at all shows that this second viscosity is of a relatively low order, for if the viscous resistance to change of

* This second viscosity appears to be of the same nature as that discussed by Shorter n his explanation of the slow extension of hair and wool fibres.† "The fibre contains elastic elements with very different degrees of damping, so that on the first application of an external force the more lightly damped elements extend, and, as time goes on, the extension of the more highly damped elements begins to show itself." And again, "We get (where a fibre is held stretched to a definite length) an apparent elastic relaxation, which, however, is very different from the effect contemplated in Maxwell's theory of viscosity. It is not the disappearance of a state of strain owing to molecular readjustments, it is merely the transference of a state of strain from lightly damped to highly damped elements."

† Shorter, S. A., 'J. Text. Inst.,' vol. T. 15, p. 207 (1924); 'Trans. Faraday Soc., vol. 20, p. 228 (1924); 'J. Soc. Dy. Col. Bradford,' vol. 41, p. 212 (1925).

shape were greater than that involved in the dissipation of internal stress, the stress would be dissipated before the dough had made any appreciable recovery. The second viscosity may be associated with the medium in which the elastic elements are embedded, but we cannot rule out the possibility of its being somehow connected with the elastic elements and their system of connection.

In relating these deductions to the known structure of the dough one may safely identify the elastic elements with the protein part of the flour. That the elements form a connected structure is confirmed by the fact that the starch and the other constituents of the flour can be washed out of the dough without breaking up the gluten. That the slowness of the elastic recovery is partly due to the presence of the starchy aqueous medium in the dough is indicated by the fact that the elastic recovery is more rapid in washed gluten than in the dough itself. The recovery of washed gluten is, however, not instantaneous, so that part of the second viscosity must be attributed to the gluten.

Our thanks are due to Professor G. I. Taylor, F.R.S., for his advice on the choice of data for inclusion in this paper, and to Dr. B. A. Keen for helpful criticism; also to Dr. E. A. Fisher, Director, and to Dr. P. Halton of the Research Association of British Flour Millers who have kindly provided the flours used in this work, and have given us much useful technical information.

Summary.

(1) An extended significance is given to Maxwell's "time of relaxation" and this has been used in quantitatively describing the viscous and elastic behaviour of flour dough.

(2) The length of the time of application of a stress in relation to the corresponding time of relaxation determines what proportion of the deformation is elastic (recoverable) and what proportion plastic (non-recoverable).

(3) This fact is illustrated by a comparison of the behaviour of dough in the "pachimeter" and on the "rack," the behaviour in the "pachimeter" (rapid stressing) being paralleled by that exhibited in a ballistic experiment.

(4) The decay of internal stress in pieces of dough which had been stretched out and held stretched has been followed, and the times of relaxation, and the corresponding viscosities have been evaluated for a series of stresses.

(5) Dough shows a phenomenon similar to the hardening of metals under working as a result of which the time of relaxation and the viscosity for a given stress depend on the total deformation.

The internal structure of the dough thus revealed is briefly considered.

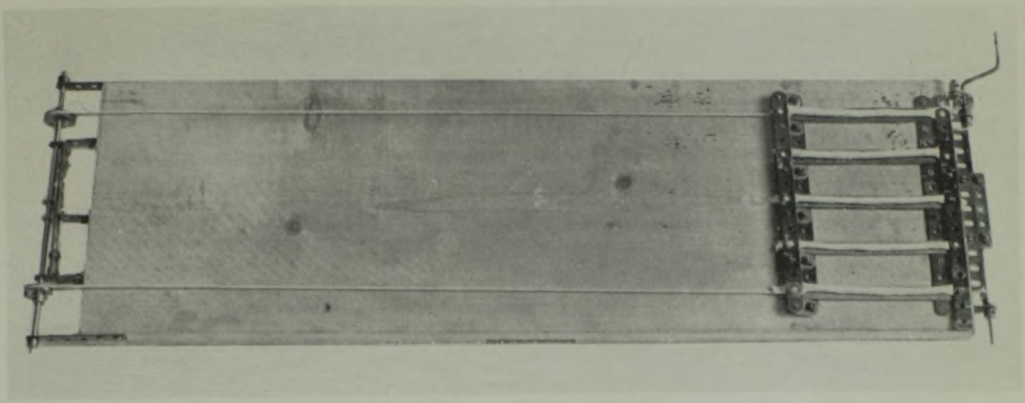

Fig. 8.

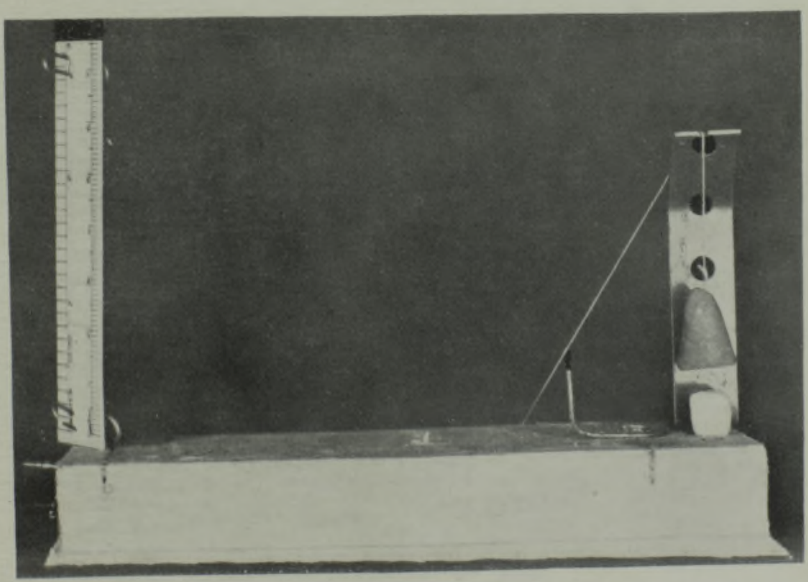

Fig. 9.